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# Radiochronological Age of a Uranium Metal Sample from an Abandoned Facility

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57

58 **Abstract**

59  
60 A piece of scrap uranium metal bar buried in the dirt floor of an old, abandoned metal rolling mill was  
61 analyzed using multi-collector inductively coupled plasma mass spectroscopy (MC-ICP-MS). The mill rolled  
62 uranium rods in the 1940s and 1950s. Samples of the contaminated dirt in which the bar was buried were also  
63 analyzed. The isotopic composition of uranium in the bar and dirt samples were both the same as natural uranium,  
64 though a few samples of dirt also contained recycled uranium; likely a result of contamination with other material  
65 rolled at the mill. The time elapsed since the uranium metal bar was last purified can be determined by the in-  
66 growth of the isotope <sup>230</sup>Th from the decay of <sup>234</sup>U, assuming that only uranium isotopes were present in the bar after  
67 purification. The age of the metal bar was determined to be 61 years at the time of this analysis and corresponds to a  
68 purification date of July 1950 ± 1.5 years.

69 **Keywords:** nuclear forensics, uranium metal, age dating, thorium, MC-ICP-MS

70 **Introduction**

71  
72 Nuclear forensics is a multidisciplinary science that uses a variety of analytical methods and tools to  
73 explore the physical, chemical, elemental, and isotopic characteristics of nuclear and radiological material in order  
74 to determine its provenance, how it was manufactured, and other factors that may affect the material characteristics.  
75 One of the important outcomes of these analyses is determining the uranium isotopic composition and when the  
76 material was last purified, *i.e.*, its radiological age. The uranium isotopic composition reveals whether the uranium  
77 is natural, depleted, or enriched and whether the material has ever been subjected to neutron irradiation and  
78 subsequently reprocessed.

79 The radiochronology (*i.e.*, age dating) presented here determines the time since purification of a uranium-  
80 bearing material based upon the ingrowth of <sup>230</sup>Th toward its parent <sup>234</sup>U in a non-disturbed environment. The  
81 uranium decay series and the position of <sup>230</sup>Th and <sup>234</sup>U in the sequence of decay products is shown in Figure 1. The

82 process of radiochronology typically employs several analytical methods that may include thermal ionization mass  
83 spectrometry (TIMS) [1-2], inductively coupled plasma mass spectrometry (ICP-MS) [3-8], gamma spectroscopy [9-  
84 11], and isotope dilution alpha spectrometry [12-13] to determine the isotopic composition and radiological age of a  
85 uranium-bearing material.

86 A small sample of a piece of uranium metal rod buried for approximately fifty years in the dirt floor of an  
87 abandoned metal rolling mill was analyzed using multi-collector inductively coupled plasma mass spectroscopy  
88 (MC-ICP-MS). The uranium metal was likely a deformed piece or an end of a longer rod that was being rolled into  
89 a uniform diameter and length for eventual use in a nuclear reactor. A suitable sample for nuclear forensic analysis  
90 was obtained from the interior of the rod to minimize the possibility of exposure to environmental factors that could  
91 alter its physical and chemical properties. The  $^{230}\text{Th}/^{234}\text{U}$  atomic ratio measured in this small (0.307 g) sample of  
92 metal was used to determine the date when the uranium metal was last purified. Samples of the dirt in which the bar  
93 was buried were also analyzed and appear to be contaminated with recycled uranium. The forensics analyses of the  
94 metal and the dirt lead to different conclusions on the origin of the uranium in these samples.

## 95 **Experimental**

### 96 *Materials*

97 All acids were from Seastar Chemical, Inc. (Sidney, BC, Canada). Poly prep chromatography columns and  
98 AG 1x8 resin bed (100-200 mesh) were from Bio-Rad Laboratories, Inc. (Hercules, CA). TEVA resin was from  
100 EiChrom Technologies, LLC (Lisle, IL). The materials used for analysis in this work included a  $^{233}\text{U}$  tracer solution  
101 calibrated with a natural uranium standard solution prepared from NBL CRM 112-A and a  $^{229}\text{Th}$  tracer solution  
102 calibrated with the NIST SRM 4342A  $^{230}\text{Th}$  radioactivity solution [3].  
103

104 The TEVA resin was prepared by repeated suspensions in a centrifuge tube containing Milli-Q water,  
105 centrifugation, and removal of the foamy later using a transfer pipet. This process removes excess organic extraction  
106 reagent and produces a more uniform size distribution of the resin particles. The AG 1x8 (100-200 mesh, chloride  
107 form) resin was prepared by repeated suspensions in Milli-Q water, allowing it to settle, and decanting and  
108 discarding any floating material. The resin was suspended twice in 6 M HCl, allowed to settle, and the acid was  
109 decanted and discarded. Finally, the resin was rinsed five times with Milli-Q water as described above.

### 110 *Sample preparation of soil*

111

112           The uranium isotopic content of three different samples of dirt were analyzed including: (1) the  
113 surrounding dirt in which the uranium metal rod was buried (raw soil), (2) the dust gently brushed from the surface  
114 of the rod (dust), and (3) yellow and white material scraped from the surface of the rod (scrapings). Samples of raw  
115 soil, dust, and scrapings were leached in nitric acid with addition of hydrofluoric acid, traced with  $^{233}\text{U}$  and  $^{229}\text{Th}$ ,  
116 and dried. The leachate containing uranium and thorium from each sample was purified and separated using a three-  
117 column procedure. First, anion-exchange chromatography was used to eliminate trace elements and organics  
118 (column A). Second, uranium was separated from the thorium using anion-exchange chromatography (column B).  
119 Third, the thorium fraction was purified using a TEVA resin bed (column C). The uranium fractions were re-  
120 dissolved in 3 mL of 2%  $\text{HNO}_3$  for isotopic uranium analysis by MC-ICP-MS. The thorium fractions were re-  
121 dissolved in a 3 mL solution of 2%  $\text{HNO}_3$  + 0.005M HF for isotopic thorium analysis by MC-ICP-MS.

#### 122 ***Radiochemical separation and purification chromatography of soil samples***

##### 123 Column A: Removal of Trace Elements

124           Trace elements and organics were removed from the leachate samples using columns containing 1.8 mL of  
125 AG1x8 (100-200 mesh) resin bed. The resin beds were pre-cleaned by rinsing with 10 mL of 0.1M HCl and 4 mL of  
126 water and then conditioned with 10 mL of 8M  $\text{HNO}_3$ . The samples (dissolved in 2 mL of 8M  $\text{HNO}_3$ ) were  
127 transferred onto the columns and the sample vials were rinsed once with 2 mL of 8M  $\text{HNO}_3$  and added to the  
128 columns. Iron was removed by rinsing the columns twice with 2 mL of 8M  $\text{HNO}_3$ . Uranium and thorium were  
129 eluted together into a 15 mL Teflon vial using 2 x 1 mL of 9M HCl, followed by 4 x 2 mL of a solution of 0.1M HCl  
130 + 0.005M HF and taken to dryness. Three drops of concentrated  $\text{HNO}_3$  followed by two drops of concentrated HCl  
131 were added to each fraction and dried.

##### 132 Column B: Uranium and Thorium Separation

133           Uranium and thorium were separated using columns containing a 1.0 mL of AG1x8 (100-200 mesh) resin  
134 bed. The columns were conditioned by rinsing with 8 mL of 0.1M HCl and 6 mL of 9M HCl. A 15 mL telfon vial  
135 was placed under each column prior to sample loading to collect the thorium fractions. Samples from column A  
136 were dissolved in 0.5 mL of 9M HCl + 10  $\mu\text{L}$  of concentrated  $\text{HNO}_3$  and loaded onto the columns, along with a  
137 single 1 mL rinse of 9M HCl from each sample container. The thorium fractions were eluted with 5 mL of 9M HCl  
138 and dried. Three drops of concentrated HCl followed by two drops of concentrated  $\text{HNO}_3$  was added to the thorium

139 vials and dried. Uranium fractions were eluted into a 15 mL Teflon vial using 7 mL of 0.1M HCl and dried. Two  
140 drops of concentrated HNO<sub>3</sub> was added twice to the uranium vials and dried.

#### 141 Column C: Thorium Purification

142 The final thorium purification step was performed using columns with 0.6 mL of TEVA resin. The  
143 columns were conditioned by rinsing with 2 mL of water and 4 mL of 4M HNO<sub>3</sub>. Thorium samples from column B  
144 were dissolved in 0.5 mL of 4M HNO<sub>3</sub> and loaded onto the columns. Sample vials were rinsed with 0.5 mL and 1.0  
145 mL of 4M HNO<sub>3</sub> and added to the columns. Residual uranium and other contaminants were removed by rinsing the  
146 columns twice with 2 mL of 4M HNO<sub>3</sub>. Thorium was eluted with 1.5 mL of 9M HCl, and 8 mL of a solution of  
147 0.1M HCl + 0.005M HF solution into 15 mL Teflon vials. After drying the thorium fractions, a series of drop  
148 additions were added to each fraction and dried down: 1) two drops of concentrated HCl 2) three drops of  
149 concentrated HCl + one drop of concentrated HNO<sub>3</sub> and 3) two drops of concentrated HNO<sub>3</sub>.

#### 150 *Sample preparation of uranium metal*

151  
152 Surface oxidation on a small piece of the uranium metal rod taken from the interior was removed by  
153 soaking in a small volume of 8M HNO<sub>3</sub> followed by a series of rinses using 8M HNO<sub>3</sub>, Milli-Q water, and then  
154 acetone. The dry, cleaned metal was weighed (0.30705 grams) and was dissolved in 4 mL of concentrated HNO<sub>3</sub> in  
155 a 30 mL Teflon vial on a hotplate until nitrogen dioxide vapors were no longer observed. A small amount of  
156 concentrated hydrofluoric acid was added to dissolve any residual precipitates. The sample was then diluted with 10  
157 mL of 8M HNO<sub>3</sub> and 12 mL of Milli-Q water. A 250 µL sample of this primary solution (0.25525 grams) was  
158 further diluted to approximately 1 liter (1003.3 grams) with 2% HNO<sub>3</sub> and called the first dilution solution. This  
159 method follows that given in Williams and Gaffney [3].

160 For the <sup>234</sup>U fraction measurement, a weighed fraction of the first dilution was spiked with <sup>233</sup>U tracer,  
161 equilibrated by heating covered on the hotplate and then dried. This fraction was re-dissolved in 3 mL of 2% HNO<sub>3</sub>  
162 and was analyzed without further purification by MC-ICP-MS. For the <sup>230</sup>Th measurement, a weighed fraction of the  
163 primary dilution was spiked with a <sup>229</sup>Th tracer and subjected to four separation and purification steps (given below)  
164 using anion-exchange resin and TEVA resin prior to analysis. The separated and purified thorium fraction was re-  
165 dissolved in 3 mL of 2% HNO<sub>3</sub> + 0.005M HF solution and analyzed by MC-ICP-MS.

#### 166 *Radiochemical separation and purification chromatography of metal thorium fractions*

167 Thorium was purified using standard techniques: initial purification was accomplished using column C  
168 (presented above); second, thorium was further purified from uranium using a 1.8 mL AG1x8 resin bed in 9 M HCl,  
169 on which uranium adsorbs and thorium passes (similar protocol to column B); third, thorium was absorbed on a 1  
170 mL AG1x8 resin bed in 8 M HNO<sub>3</sub> and then eluted with 9 M HCl followed by 0.1 M HCl + 0.005 M HF (similar  
171 protocol to column A); and lastly, the thorium was passed through a 1 mL AG1x8 resin bed in 9 M HCl.

## 172 ***Instrumentation***

### 173 *MC-ICP-MS*

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176 The uranium and thorium mass spectrometric analyses were performed using a NuPlasma HR multi-  
177 collector ICPMS with a combination of Faraday and electron multiplier (pulse-counting) detectors. Samples were  
178 introduced to the plasma via a CETAC Aridus II system with a 100 mL/min Teflon nebulizer. Instrumental mass  
179 bias and detector cross-calibration factors for both the U and Th analyses were determined using a certified  
180 reference material (CRM U010) of 1% enriched uranium obtained from the U. S. Department of Energy New  
181 Brunswick Laboratory. The isotope dilution tracers, <sup>233</sup>U for uranium and <sup>229</sup>Th for thorium, were calibrated with  
182 gravimetrically prepared standard solutions of U metal (NBL CRM 112-A) and NIST SRM 4342A <sup>230</sup>Th  
183 radioactivity solution. Thorium was measured by peak-hopping <sup>230</sup>Th-<sup>229</sup>Th on an electron multiplier with  
184 simultaneous analysis of <sup>232</sup>Th on Faraday cups. Uranium was measured in static multi-collection mode with <sup>234</sup>U  
185 and <sup>233</sup>U on electron multipliers and <sup>235</sup>U and <sup>238</sup>U on Faradays. Memory effects were corrected by measuring blank  
186 acid solutions that were used to dissolve the purified samples immediately prior to sample analysis. Data reduction  
187 involved correcting for detector baseline, acid blank, detector cross-calibration factor, and instrumental mass bias.

## 188 **Results and Discussion**

189 The uranium isotopic composition determined for each of the samples analyzed is listed in Table 1 and is  
190 similar to that of natural uranium (i.e., <sup>238</sup>U = 99.2745 ± 0.0015%; <sup>235</sup>U = 0.7200 ± 0.0012%; <sup>234</sup>U = 0.0055 ±  
191 0.0005%) [14]. The lack of measureable <sup>236</sup>U in the uranium metal and the scraping samples indicates that these  
192 samples had not been subjected to nuclear fuel reprocessing or recycling. The presence of a small quantity of <sup>236</sup>U  
193 measured in the dust and raw soil samples may reflect contamination from recycled uranium associated with other  
194 work at the facility. Table 2 lists the total uranium mass and activity concentration determined for each sample.  
195 The piece of metal is 98.37% natural uranium and reflects a composition expected for uranium metal from this era.

196 The yellow and white material scraped from the metal bar are likely uranium metal oxides that were formed on the  
 197 surface of the metal bar while it was buried for over fifty years and exposed to ambient environmental conditions.

198 The ratio of  $^{235}\text{U}/^{238}\text{U}$  determined for each sample is shown on Figure 2 where the solid line indicates the  
 199 ratio for natural uranium (0.0072527) and the dashed lines indicate +/- 0.08% [15]. The low ratio observed for the  
 200 raw soil may suggest that there is some contamination from depleted uranium, which had also been rolled at the  
 201 facility. The uranium contamination in the dirt floor is very heterogeneous and probably reflects the different types  
 202 of metals that were rolled at the facility over time.

203 The calculated radiological age of each sample is listed on Figure 3 and has been determined from in-  
 204 growth of  $^{230}\text{Th}$  from radioactive decay of  $^{234}\text{U}$  with the assumptions that (1) no  $^{230}\text{Th}$  was present at the time the  
 205 uranium metal was last purified, and (2) no thorium contamination was introduced after uranium purification. The  
 206 mathematical expression for determining the time (t) since last purification, based upon the atom ratio of  $^{230}\text{Th}$  to  
 207  $^{234}\text{U}$  is:

$$208 \quad t = \frac{1}{(\lambda_{234} - \lambda_{230})} \times \ln \left[ 1 + \frac{N_{230\text{Th}}}{N_{234\text{U}}} \times \frac{(\lambda_{234} - \lambda_{230})}{\lambda_{234}} \right]$$

209 where  $\lambda_{234}$ ,  $\lambda_{230}$ ,  $N_{234\text{U}}$ , and  $N_{230\text{Th}}$  are the decay constants and number of atoms for  $^{234}\text{U}$  and  $^{230}\text{Th}$ , respectively. The  
 210 number of atoms of uranium and thorium were determined from the isotope dilution MC-ICP-MS analysis. The  
 211 half-lives used for  $^{234}\text{U}$  and  $^{230}\text{Th}$  are  $245,250 \pm 490$  years ( $2\sigma$ ) and  $75,690 \pm 230$  years ( $2\sigma$ ), respectively [16].

212 The relative age of uranium metal and the scraping are approximately the same (Fig. 3). The calculated age  
 213 of the uranium metal is 61 years, corresponding to a purification date of July of  $1950 \pm 1.5$  years. The calculated  
 214 age of the scrapings is 60 years, corresponding to August of  $1951 \pm 3.5$  years. The radiological age of the raw soil  
 215 and dust is significantly less than the metal or scrapings and may be an artifact of the heterogeneity of the uranium  
 216 contamination in the dirt floor, mobility of thorium and uranium due to environmental transport, or housekeeping  
 217 practices during plant operation that diluted or removed contamination in the floor.

### 218 Conclusions

219 Radiochronology methods were used to analyze samples from an abandoned site in order to ascertain the  
 220 material and isotopic composition and radiological age of suspected uranium-bearing materials. The uranium  
 221 isotopic compositions measured in each sample is similar to that of natural uranium other than the slight  $^{236}\text{U}$   
 222 detected in the dust and raw soil samples. The radiological age calculated for the raw soil and dust is significantly

223 less than the samples of metal and scrapings. The difference in radiological age may be attributed to environmental  
224 transport or the results of clean-up activities to mitigate contamination while the facility was in operation. The  
225 findings from these analyses are evident that methods employed in radiochronology are valuable in ascertaining the  
226 provenance of intercepted materials. The age determination of the uranium metal was calculated to be 61 years  
227 corresponding to a production date of July of 1950  $\pm$  1.5 years. The uranium metal is better suited for forensic  
228 analysis, as it is not as subject to the leaching and other factors which may fractionate thorium and uranium in soil  
229 samples.

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Table 1. Uranium isotopic composition in samples

Sample Description	<sup>238</sup> U, atom %	<sup>236</sup> U, atom % <sup>A</sup>	<sup>235</sup> U, atom %	<sup>234</sup> U, atom %
U Metal Scrapings	99.27 ± 0.12	None detected	0.7206 ± 0.0006	0.00541 ± 1.7·10 <sup>-5</sup>
U Metal Dust	99.27 ± 0.12	1.68·10 <sup>-6</sup> ± 5.3·10 <sup>-7</sup>	0.7200 ± 0.0006	0.00541 ± 1.7·10 <sup>-5</sup>
Raw Soil	99.28 ± 0.12	2.33·10 <sup>-5</sup> ± 5.5·10 <sup>-7</sup>	0.7176 ± 0.0006	0.00538 ± 1.7·10 <sup>-5</sup>
U Metal	99.27 ± 0.14	None detected	0.7200 ± 0.0013	0.005412 ± 1.7·10 <sup>-5</sup>
Natural Uranium <sup>B</sup>	99.27 ± 0.0015	0	0.7200 ± 0.0012	0.005505 ± 0.0005

274 \*U uncertainties are given as the combined standard uncertainty.

275 <sup>A</sup> <sup>236</sup>U was determined to contribute less than 1E-6 atom percent based on the detection limit.

276 <sup>B</sup> Corresponds to reference 14.  
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*Table 2.* Total uranium concentration in samples

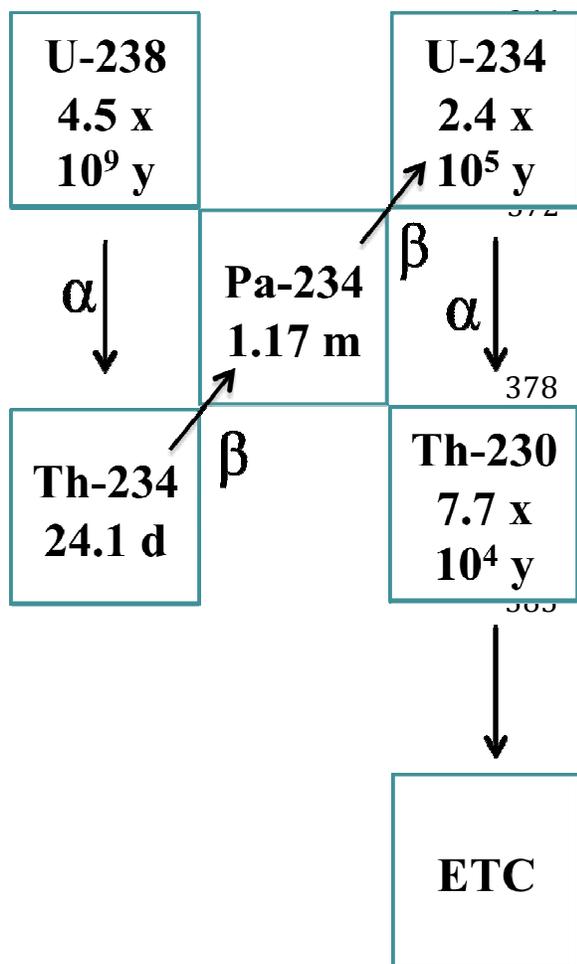
<b>Sample Description</b>	<b>U, g/g</b>	<b>μCi/g</b>
U Metal Scrapings	$0.586 \pm 0.011$	$0.3988 \pm 0.0074$
U Metal Dust	$0.257 \pm 0.005$	$0.1748 \pm 0.0036$
Raw Soil	$0.048 \pm 0.001$	$0.0330 \pm 0.0007$
U Metal	$0.984 \pm 0.005$	$0.6696 \pm 0.0035$

\*All uncertainties are the combined standard uncertainty.

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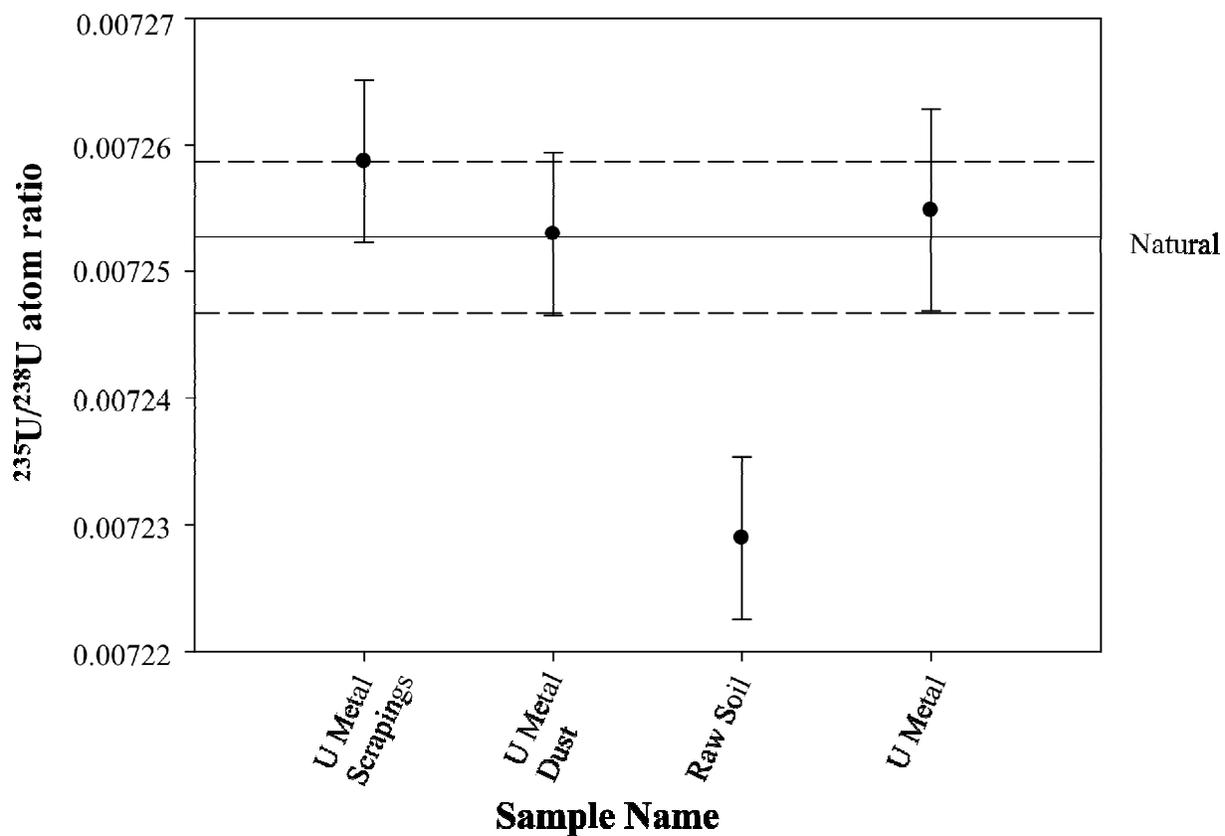
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Figure 1: <sup>238</sup>U decay chain series displays the decay of <sup>234</sup>U into <sup>230</sup>Th.

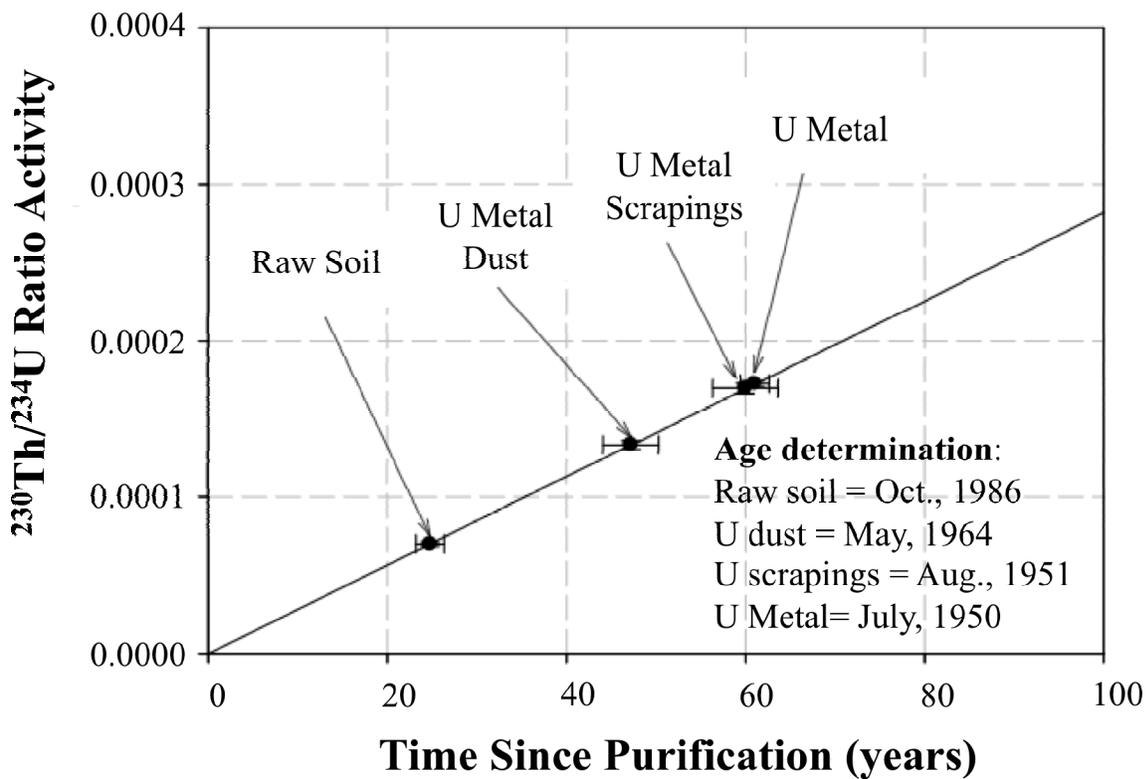
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**Figure 2:** Soil and uranium metal sample  $^{235}\text{U}/^{238}\text{U}$  ratio comparison. The solid line indicates the ratio for natural uranium (0.0072527) and the dashed lines indicate +/- 0.08% [15]. Error bars are the combined standard uncertainty.

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Figure 3: Age determination comparison of each sample. Error bars are expanded uncertainties ( $k=3$ ).